

Qiang Xue
 Jiang-Shan Li
 Ping Wang
 Lei Liu
 Zhen-Ze Li

State Key Laboratory of
 Geomechanics and Geotechnical
 Engineering, Institute of Rock and
 Soil Mechanics, Chinese Academy of
 Sciences, Hubei, P. R. China

Research Article

Removal of Heavy Metals from Landfill Leachate Using Municipal Solid Waste Incineration Fly Ash as Adsorbent

To study the adsorption effect of municipal solid waste incineration (MSWI) fly ash on removal of heavy metals from landfill leachate, batch tests were performed to study the effects of MSWI fly ash dosage, pH of leachate, and reaction time on adsorption. The adsorption mechanism of MSWI fly ash to heavy metals was analyzed through scanning electron microscope and X-ray diffraction tests. With fly ash dosage of 10 g/L, leachate pH 9, and reaction time of 60 min, the removal rate of Zn, Pb, Cr, Cd, and Cu in the leachate reached 39.42, 59.24, 28.14, 55.37, and 32.82%, respectively. The pseudo first-order kinetic equation could well fit the adsorption kinetic process. The adsorption mechanisms of MSWI fly ash to heavy metals in the leachate mainly included ion exchange adsorption, electrostatic adsorption, and chemical fixation.

Keywords: Activated carbon; Adsorption; Modeling; MSWI fly ash; Sewage pollutants

Received: August 19, 2013; *revised:* November 17, 2013; *accepted:* December 9, 2013

DOI: 10.1002/clean.201300619

1 Introduction

Leachate from landfills contains abundant, highly concentrated heavy metals [1]. The nitrogen and organic pollutants in leachate can be degraded because such pollutants can easily undergo physical, chemical, and biological reactions. However, the heavy metals in the leachate cannot be decomposed by the microorganisms in the soil layer. However, these metals can be concentrated by organisms and have strong mobility, causing pollution to soil and ground water. Completely removing heavy metals from soils is difficult, thus posing a serious threat to human health [2, 3]. The traditional treatment method for heavy metals in leachate mainly involves the physico-chemical methods, including adsorption, chemical precipitation, and catalytic oxidation [4–6]. Research on the treatment of sewage pollutants using low-cost adsorbents has recently attracted considerable attentions [7–9]. Milicevic et al. studied the removal effect of kolubara lignite on copper from aqueous solutions, which revealed that about 90% of copper cations were removed in 5 min of contact time from the solution with the lowest copper concentration (50 mg Cu²⁺/L) regardless adsorbent amount [10]. Research on the adsorption effect of activated carbon on heavy metal ions indicated that the pH value and activated carbon dosage were the main influence factors, and the order of absorptive capacity was Fe²⁺ > Cu²⁺ > Ni²⁺ [11]. Issabayeva et al. removed copper ions from aqueous solutions using palm shell activated carbon as an adsorbent [12]. Research results showed that 98% of Pb in an

aqueous solution could be absorbed by bentonite [13]. Municipal solid waste incineration (MSWI) fly ash refers to the mixture of slag and stove ash produced during the burning of refuse, the quality of which is approximately 5% of the waste. MSWI fly ash is a toxic waste with abundant heavy metals and dioxin absorbed from smoke. Safe disposal of MSWI fly ash, as well as its resource utilization technique, has attracted considerable attention from domestic and foreign scholars [14, 15]. MSWI fly ash is an efficient adsorbent, because it has small particles but large pores and a specific surface area with pH > 10 [16]. No systematic research has been conducted on the application of MSWI fly ash for the adsorption of heavy metals in wastewater, leaving its associated control conditions and reaction mechanism unclear.

In this paper, the adsorption characteristics of MSWI fly ash on five heavy metals in leachate were studied through a series of laboratory experiments, which demonstrated the effects of MSWI fly-ash dosage, reaction time, and pH value of leachate on the removal rate of heavy metals. We also proposed an optimal mathematical model of adsorption kinetics and revealed the adsorption reaction mechanism based on microcosmic tests.

2 Materials and methods

2.1 MSWI fly ash

MSWI fly ash from the Likeng refuse incineration plant, Guangzhou, China, was sampled. The fly ash was dried at 105°C and screened using a 0.25 mm soil sieve. The screened fly ash was then sealed up for application. The chemical components of the MSWI fly ash were determined by an Axios advanced X-ray fluorescence spectrometer from PANalytical, The Netherlands. The main mineral components of the fly ash were measured by using X-ray diffractometry (XRD, D8 Advance) (Tab. 1). The prepared fly ash samples were exposed to X-ray ($k = 1.54060 \text{ \AA}$) with the 2θ angle varying between 10° and 80° with Cu K α radiation. The applied voltage and current were 40 kV and

Correspondence: Professor Q. Xue, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, 430071, Bayi Road, WuChang District, Wuhan City, Hubei Province, P. R. China
 E-mail: qiangx@whrsm.ac.cn

Abbreviations: ICP-OES, inductively coupled plasma optical emission spectrometry; MSWI, municipal solid waste incineration; PZC, point of zero charge; SEM, scanning electron microscopy; XRD, X-ray diffractometry

Table 1. The main mineral components of tested MSWI fly ash before and after adsorption

| Composition | Content (%) | |
|-------------|-------------|--------------------------|
| | Raw fly ash | Fly ash after adsorption |
| Sylvite | 32.00 | 0.14 |
| Anhydrite | 30.65 | 40.88 |
| Quartz | 10.01 | 4.7 |
| Anorthite | 1.41 | 1.87 |
| Halite | 5.98 | 0.64 |
| Gismondine | 12.92 | 10.72 |
| Calcite | 3.22 | 36.71 |
| Hematite | 3.81 | 4.32 |

30 mA, respectively. The microstructure of MSWI fly ash after fully saturated with deionized water was observed by using scanning electron microscopy (SEM, Quanta 250) (Fig. 1). The pH was determined by mixing 2 g of fly ash with 100 mL of distilled water and was then recorded at 1 h intervals for 24 h. The specific surface area of the fly ash was determined by using the Brunauer–Emmett–Teller method. The point of zero charge (pzc) was determined using the solid addition method [17]. The experimental results, Tab. 2, show that MSWI fly ash has large specific surface area, a high alkalinity, and contains abundant metallic oxides (SiO₂ and Al₂O₃ contents make up about 17.44% of the fly ash, while Fe₂O₃ and CaO compose about 46.68%), thus making it an efficient adsorbent material.

The leaching concentration of the heavy metals in the fly ash was tested by using the toxicity characteristic leaching procedure. First, 20 g fly ash were placed in a 2 L extraction flask, after which 400 mL acetum (obtained by diluting 17.25 mL pure glacial acetic acid to 1 L with distilled water) were added. The extraction flask was fixed onto the turnover oscillator, the rotation speed of which was adjusted to 30 ± 2 rpm. The mixed solution was oscillated for 18 ± 2 h at 23 ± 2 °C and was filtered using vacuum filtration equipment. The heavy metals concentration in the filtered solution was measured by using inductively coupled plasma optical emission spectrometry (ICP-OES) (Optima 4300DV, PerkinElmer, USA) as shown in Tab. 3.

2.2 Landfill leachate

The landfill leachate used in the tests was taken from the Chengjiachong refuse landfill, Wuhan City, China, and its water

Table 2. Chemical and physical properties of sieved MSWI fly ash

| Property | Value | Property | Value |
|------------------------------------|-------|-------------------------------------------|-------|
| CaO (%) | 44.31 | CO ₂ (%) | 2.89 |
| SO ₃ (%) | 8.25 | K ₂ O (%) | 4.13 |
| SiO ₂ (%) | 11.00 | Fe ₂ O ₃ (%) | 2.37 |
| Na ₂ O (%) | 4.77 | pH | 10.82 |
| Al ₂ O ₃ (%) | 6.44 | pH _{pzc} | 6.13 |
| MgO (%) | 1.62 | Specific surface area (m ² /g) | 9.475 |

quality indexes are shown in Tab. 4. The pH of the leachate was measured by a PHSJ-4A pH meter from Shanghai Precision & Scientific Instruments, and the heavy metals concentrations were measured by ICP-OES. In addition, chemical oxygen demand was measured by permanganate titration, and NH₃-N was measured by Nessler’s reagent spectrophotometry.

2.3 Experimental methods

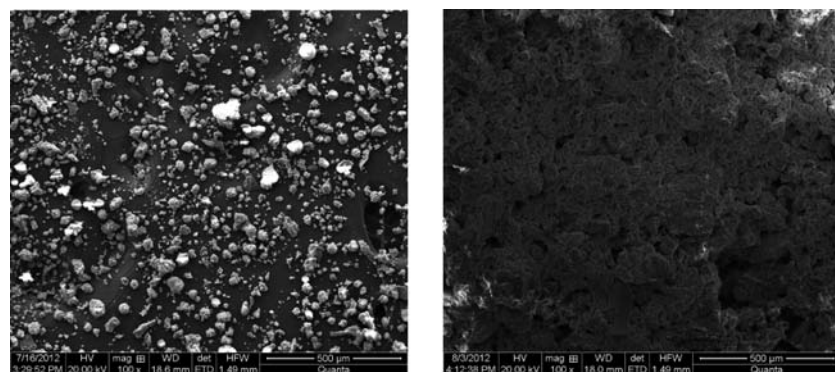
Batch adsorption experiments were carried out. 100 mL leachate were weighed and poured in a series of 150 mL conical flasks. Certain quantity of MSWI fly ash (0.1, 0.25, 0.5, 1, 1.5, and 2 g) was weighed and added into conical flasks to obtain MSWI fly ash slurry. The pH of the slurry was adjusted to the desired value in the range of pH 3–11 with 0.1 M HCl and 0.1 M NaOH. The conical flasks were placed in a thermostatic water bath vibrator and oscillated at 25 °C and 150 rpm for different times (5, 15, 30, 60, 90, 120, 180, and 240 min) in the kinetic studies. At the end of agitation period the mixture was centrifuged for 10 min at 6000 rpm. The concentration of heavy metals in the supernatant was measured using ICP-OES. Each test was conducted in triplicate, the reported value of heavy-metals ions adsorbed by fly ash in each test were the average of three measurements.

The removal rate of heavy-metals ions adsorbed by MSWI fly ash was taken as the difference between the initial and residual concentration of the heavy-metals ions. The calculation formula is:

$$\beta(\%) = \frac{(C_0 - C_1)}{C_0} \times 100 \quad (1)$$

where C₀ is the initial concentration of the heavy metal-ions in the leachate (mg/L), and C₁ is the residual concentration of the heavy-metal ions in the leachate after adsorption by MSWI fly ash (mg/L).

To study the adsorption mechanisms of MSWI fly ash. The centrifugal-filtered fly ash was dried at 105 °C. The scanning electron



(a) Raw MSWI fly ash

(b) MSWI fly ash after adsorption

Figure 1. Scanning electron micrographs of MSWI fly ash before and after adsorption.

Table 3. Leached heavy metals concentration in fly ash

| Heavy metal | Leaching concentration (mg/L) |
|-------------|-------------------------------|
| Pb | 3.521 |
| Cd | 0.964 |
| As | 2.819 |
| Ni | 0.190 |
| Cu | 0.672 |
| Hg | 0.428 |
| Zn | 28.300 |
| Cr | 1.847 |

Table 4. Characteristics of landfill leachate

| Parameter | Value |
|--------------------|--------|
| pH | 7.48 |
| COD | 5625.5 |
| NH ₃ -N | 66.8 |
| Pb | 0.355 |
| Zn | 0.266 |
| Cr | 0.112 |
| Cd | 0.207 |
| Cu | 0.209 |

All parameters are expressed in mg/L except pH. COD, chemical oxygen demand.

micrograph after fully saturated with deionized water was measured by SEM. The dried fly ash was ground and passed a 200 mesh sieve, and the mineral components were measured with XRD.

3 Results and discussion

3.1 Effect of fly ash dosage on the adsorption of heavy metals in leachate

The effect of adsorbent dosage on the removal rate of heavy metals in leachate was analyzed by varying the fly ash dosage from 1 to 20 g/L. The pH value of the leachate was 9, and the reaction time was 60 min. The experimental results are shown in Fig. 2, which showed

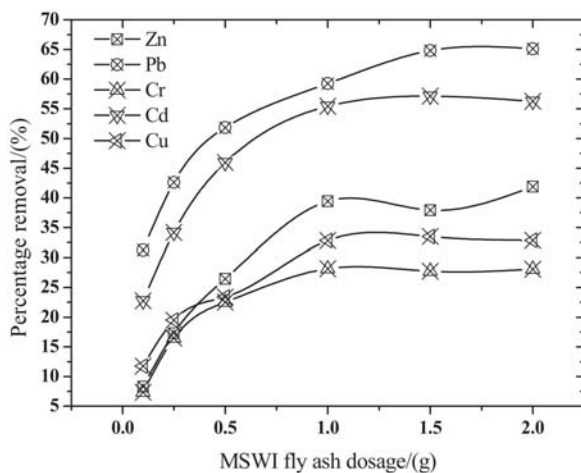


Figure 2. Effect of MSWI fly ash dosage on percentage removal of heavy metals in landfill leachate.

that with the increase in fly ash dosage (from 1 to 10 g/L), the removal rate of the heavy metals in the leachate increased continuously by 31.14, 27.97, 20.79, 32.66, and 21.09% for Zn, Pb, Cr, Cd, and Cu, respectively. The removal capacities of fly ash to heavy metals in the leachate were in the order Pb > Cd > Zn > Cu > Cr. However, the removal rate of heavy metals tended to balance thereafter, which indicated that the optimal dosage of fly ash was approximately 10 g/L, and the adsorption sites provided by the adsorbent attained saturation at this dosage. Amount of heavy metals adsorbed per gram of the MSWI decreased. This is probably due to the availability of more adsorbent surface for adsorbate species.

3.2 Effect of reaction time on the adsorption of heavy metals in leachate

The effect of reaction time on the adsorption of heavy metals in leachate is shown in Fig. 3. The pH value of the leachate was 9, and the fly-ash dosage was fixed at 10 g/L. As shown in the figure, with the extension in adsorption time, the removal rate of the heavy metals achieved a sharp increase initially and then gradually increased until a plateau appeared, which indicated a dynamic balance. The instant adsorption rate decreased gradually to zero, and the balance point of adsorption lay between 60 and 90 min. The adsorption reaction can be divided into the quick adsorption, slow adsorption, and dynamic adsorption balance periods. In the initial adsorption period, abundant and unoccupied adsorption sites existed on the adsorbent surface. Thus, the adsorption rate was rapid. With prolonged time, the adsorption sites were gradually occupied by heavy-metal ions. The electrostatic repulsion between the metal ions in the adsorption phase and those in the solution phase was strengthened, and the concentration gradient (impetus) for the solutes to spread to the adsorbent surface decreased, thus resulting in adsorption difficulty in the later period.

3.3 Effect of pH of the leachate on the adsorption of heavy metals

The pH of the leachate was varied from 3 to 11 to study its effect on the adsorption of heavy metals by fly ash. The experimental results

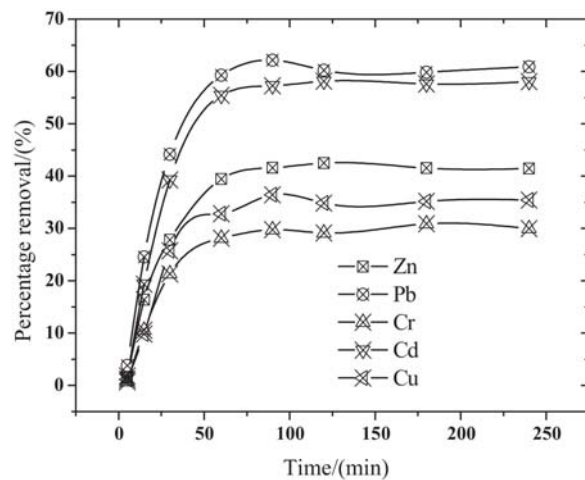


Figure 3. Effect of contact time on percentage removal of heavy metals in landfill leachate.

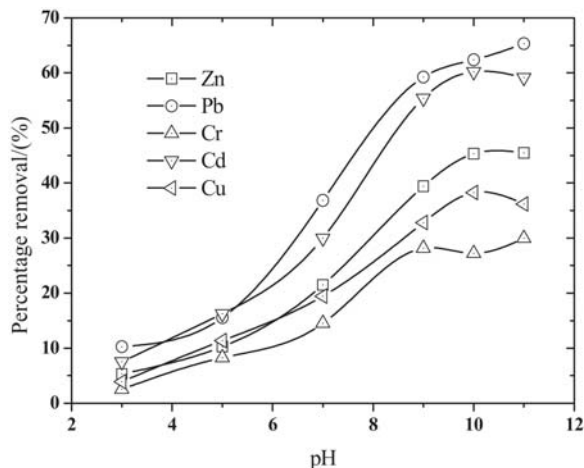


Figure 4. Effect of pH on percentage removal of heavy metals in landfill leachate.

are shown in Fig. 4, which indicated that the pH exerted a significant effect on the removal rate of heavy metals because the pH value of the solution determined the charge on the absorbent surface [18]. When the pH of leachate was small, the removal rate of heavy metals in the leachate was low. The experiment indicated that at pH 3, the removal rate of some heavy metals was negative (the values shown in Fig. 4 are the average value of three results) because some soluble heavy metals were dissolved in the leachate by strong acid. With the increase in pH of the leachate (from 3 to 10), the removal rate of the heavy metals increased continuously by 40.00, 52.12, 24.69, 52.67, and 34.32% for Zn, Pb, Cr, Cd, and Cu, respectively. When the pH value increased from 7 to 9, the removal rate of the heavy metal increased rapidly, which indicated that alkaline environment could facilitate the removal rate of heavy metals. However, when the pH value increased further, the removal rate of the heavy metals showed no significant change, and that of some heavy metals even decreased because some heavy-metal precipitations formed by adsorption again dissolved into the leachate through a hydration reaction [19].

The effect of pH of the leachate on the removal rate of heavy metals by MSWI fly ash can be explained by the pH_{pzc} . When the solution $pH < pH_{pzc}$, the fly-ash surface is positively charged, whereas when the solution $pH > pH_{pzc}$, the fly-ash surface is negatively charged. The pH_{pzc} of fly ash sample is 6.13 and a major change in adsorption was observed at pH 6–7 for all five metal ions studied. Heavy-metal ions are difficultly absorbed by the positively charged fly-ash surface for the existence of repulsive force when pH of leachate is low. Meanwhile, H^+ in leachate can compete with heavy-metal ions for the adsorption, which leads to the low removal rate of heavy metals. When $pH > pH_{pzc}$, heavy-metal ions and metal hydroxides can be easily absorbed by the negatively charged fly-ash surface. A number of studies indicated that hydroxyl metal complex is more easily absorbed than hydrated metals because the OH group of metal ions decreases the free energy necessary for adsorption [20, 21].

3.4 Adsorption kinetics

The adsorption process of heavy metals by MSWI fly ash was simulated using three widely applied kinetic models: pseudo first-order kinetics, pseudo second-order kinetics, and particle internal diffusion. The

adsorption kinetics can not only deduce the adsorption mechanism through the speed rate but can also determine the time required for the adsorption to achieve balance [22, 23].

The pseudo first-order kinetics model is described by Eq. (2):

$$\lg(q_e - q_t) = \lg q_e - \frac{k_1}{2.303} t \quad (2)$$

where q_e and q_t (mg/g) are the absorbed amounts of solute per unit quality of absorbent at the balanced moment and at t (min), and k_1 (min^{-1}) is the adsorption speed constant of the pseudo first-order kinetics.

The pseudo second-order kinetic equation can be expressed as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (3)$$

where k_2 (g/mg/min) is the adsorption speed constant of the pseudo second-order kinetics.

The simplified particle internal diffusion equation is [24]:

$$q_t = k_{int} t^{0.5} + C \quad (4)$$

The fitting curves of adsorption process for the five heavy metals on MSWI fly ash by the pseudo first-order kinetics, pseudo second-order kinetics, and particle internal diffusion models are shown in Fig. 5. The fitted adsorption kinetics parameters are shown in Tab. 5. The related coefficient of fitting results showed that the pseudo first-order kinetics model was the most appropriate model to describe the adsorption process of the five heavy metals in leachate on MSWI fly ash. This result indicated that the adsorption process of several heavy metals on the fly ash was mainly influenced by the diffusion resistance of liquid films [25]. With respect to Zn, Pb, Cr, Cd, and Cu, their adsorption amounts predicted by the pseudo first-order kinetics model were 1.138, 2.205, 0.345, 1.232, and 0.762 mg/g, respectively, which agreed well with the test results.

The rate constant of the pseudo second-order kinetics for the five heavy metals showed no significant change, which indicated the adsorption speed rate of the various heavy metals and the time for the adsorption reaction of the fly ash and heavy metals in the leachate to achieve balance were similar. Among the related coefficients of the five heavy metals, only Pb showed 0.9, whereas the rest achieved small values. The formation of chemical bond was the main factor that influenced the adsorption effect of the pseudo second-order kinetics [26]. This result indicated that the adsorption process of fly ash and heavy metals was not determined by chemical adsorption.

If the fitting result of the particle internal diffusion model shows a linear relationship, the adsorption process is only controlled by the particle internal diffusion; otherwise, the adsorption process is controlled by two or more steps [24]. The fitting result of the particle internal diffusion model for the five heavy metal ions showed a weak linear relationship during the entire adsorption period (the related coefficient value was between 0.6 and 0.7), which indicated that the adsorption process had several rate-control steps. Therefore, by comparing comprehensively the fitting results of the three models, the adsorption mechanism of MSWI fly ash to heavy metals in the leachate was mainly controlled by the liquid film diffusion, accompanied with the effect of particle internal diffusion.

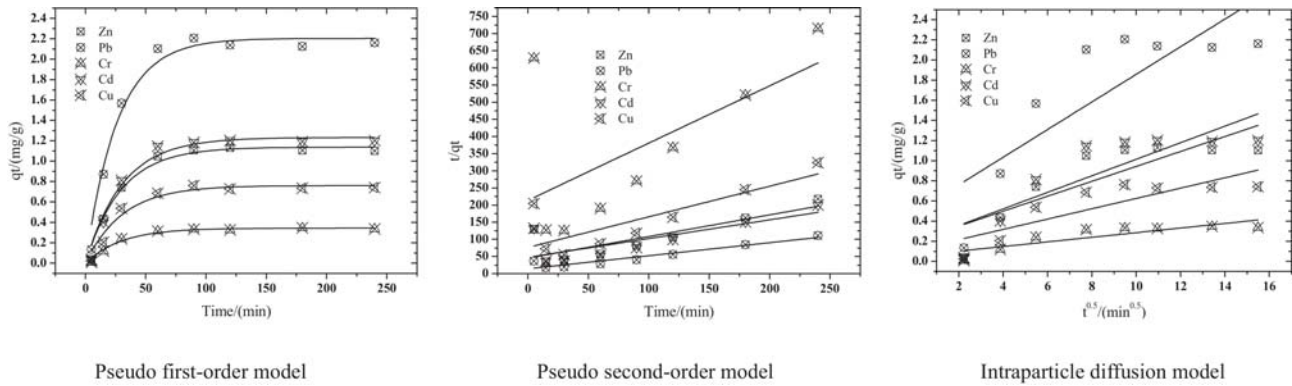


Figure 5. Adsorption kinetics curves of heavy metals in landfill leachate by MSWI fly ash.

Table 5. Adsorption kinetic model rate constants for heavy metals

| Heavy metal | Pseudo first-order | | | Pseudo second-order | | | Intraparticle diffusion model | | |
|-------------|--------------------|-------------------------------------------|-------|---------------------|--------------------------------|-------|----------------------------------------------|-------|-------|
| | q_e (mg/g) | Rate constant k_1 (min^{-1}) | R^2 | q_e (mg/g) | Rate constant k_2 (g/mg min) | R^2 | k_{int} ($\text{mg/g min}^{0.5}$) | c | R^2 |
| Zn | 1.138 | 0.034 | 0.974 | 1.586 | 0.009 | 0.625 | 0.074 | 0.202 | 0.669 |
| Pb | 2.205 | 0.038 | 0.971 | 2.631 | 0.010 | 0.900 | 0.137 | 0.485 | 0.631 |
| Cr | 0.345 | 0.036 | 0.965 | 0.599 | 0.013 | 0.263 | 0.023 | 0.056 | 0.676 |
| Cd | 1.232 | 0.033 | 0.967 | 1.827 | 0.006 | 0.534 | 0.082 | 0.192 | 0.673 |
| Cu | 0.762 | 0.032 | 0.950 | 1.120 | 0.010 | 0.571 | 0.051 | 0.112 | 0.656 |

3.5 Adsorption mechanism

SEM and XRD tests of MSWI fly ash before and after adsorption were conducted to study the adsorption mechanism of MSWI fly ash to the heavy metals in leachate. The test results are shown in Fig. 1 and Tab. 1. The results showed that MSWI fly ash was full of fine particles and spaces before the adsorption. However, after the adsorption, honeycomb was appeared on the surface of fly ash with abundant leachate pollutants covering it. The spaces in the fly ash were filled gradually. This result indicated that fly ash underwent physical adsorption to the leachate because fly ash has strong adsorption capacity attributed to its large specific surface area [27]. So the adsorption process of fly ash and heavy metals was determined by physical adsorption (electrostatic attraction), which was also obtained by Papandreou et al. [28]. Table 1 shows that the mineral components of the fly ash changed significantly before after the adsorption, among which, the kali salt (main component is KCl) decreased to 0.14 from 32%, and the rock salt (main component is NaCl) decreased to 0.64 from 5.98%. Therefore, we can deduce that the sodium and kali salts in the fly ash underwent ion exchange and dissolution reactions. Abundant K^+ and Na^+ in the fly ash were replaced or dissolved into the leachate. Quartz (main component is SiO_2) decreased to 4.7 from 10.01% because of the strong adsorption capacity of SiO_2 toward heavy metals [29]. At $\text{pH} > 6.13$, a negatively charged silica surface appeared, on which a large amount of heavy metals and oxy-hydrogen were synthesized [30]. On the surface of the MSWI fly ash the functional oxidized groups are present as SiO_2 , Fe_2O_3 , and Al_2O_3 , alumina and iron also showed the same phenomenon of developing negative charges depending on pH. Calcite (main component is CaCO_3) increased from 3.22 to 36.71% because the fly ash and the atmosphere contain CO_2 , which easily forms CO_3^{2-} under an alkaline environment. Plenty of CaCO_3 was

produced for the fly ash contains a large amount of Ca^{2+} . Gypsum (main component is CaSO_4) content increased from 30.65 to 40.88% because the SO_3 in fly ash reacted with water to produce SO_4^{2-} .

4 Conclusions

Although MSWI fly ash is a hazardous waste, this paper verified through a series of experiments that it can be used as an efficient absorbent for the adsorption of heavy metals in landfill leachate for the characteristic of fine particle, high pH and high concentration of active oxide. The MSWI fly ash covered with adsorbed heavy metals can be disposed in landfill after stabilization/solidification treatment [31]. Furthermore, the adsorption mechanism of MSWI fly ash to heavy metals was revealed through microscopic tests. The following conclusions were obtained:

- (i) MSWI fly ash has a large specific surface area as well as high pH value and contains abundant metallic oxides, thus making it an efficient absorbent. The optimal dosage of MSWI fly ash for the adsorption of heavy metals in leachate was approximately 10 g/L, with which the adsorption sites attained saturation. The removal capacity of fly ash to heavy metals in the leachate was in the order of $\text{Pb} > \text{Cd} > \text{Zn} > \text{Cu} > \text{Cr}$.
- (ii) The equilibrium time of adsorption reaction between the MSWI fly ash and heavy metals in the leachate was within 60–90 min. The process of adsorption reaction could be divided into the quick adsorption, slow adsorption, and dynamic adsorption balance periods. The pseudo first-order kinetics model was the most appropriate model for describing the adsorption process of heavy metals in leachate by MSWI fly ash. The adsorption mechanism was mainly

controlled by liquid film diffusion, accompanied with the effect of particle internal diffusion.

- (iii) An alkaline environment can facilitate the removal of heavy metals in leachate by MSWI fly ash. When the pH of landfill leachate increased from 7 to 9, the removal rate of heavy metal increased rapidly. The pH value of the leachate determines the charge on the surface of MSWI fly ash. When $\text{pH} > \text{pH}_{\text{PZC}}$, heavy metal ions and their precipitation of hydroxides can easily be absorbed by the negatively charged fly-ash surface.
- (iv) The scanning electron micrographs of MSWI fly ash before and after adsorption indicated that the fly-ash surface absorbed abundant pollutants from the landfill leachate. Analyzed by combining the XRD results, it was concluded that the adsorption mechanism of fly ash on heavy metals in leachate mainly included electrostatic adsorption, ion exchange, and chemical fixation.

Acknowledgements

This research was supported by the National Basic Research Program of China (973 Program) (2012CB719802), the National Water Pollution Control and Management Science and Technology Major Projects of China (2012ZX07104-002), Wuhan high and new technology achievements transformation and industrialization project (2013060803010403).

The authors have declared no conflict of interest.

References

- Y. Z. Peng, S. J. Zhang, S. W. Zheng, S. Y. Wang, Heavy Metal Ions in the Biological Treatment of Municipal Landfill Leachate, *Tech. Equip. Environ. Pollut. Control* **2006**, 7 (1), 1–5.
- D. L. Baun, T. H. Christensen, Speciation of Heavy Metals in Landfill Leachate: A Review, *Waste Manage. Res.* **2004**, 22 (1), 3–23.
- B. Nowak, S. Frias Rocha, P. Aschenbrenner, H. Rechberger, F. Winter, Heavy Metal Removal from MSW Fly Ash by Means of Chlorination and Thermal Treatment: Influence of the Chloride Type, *Chem. Eng. J.* **2012**, 179 (1), 178–185.
- Q. Wei, Z. Ying, Removal of Lead, Copper, Nickel, Cobalt, and Zinc from Water by a Cancrinite-type Zeolite Synthesized from Fly Ash, *Chem. Eng. J.* **2009**, 145 (3), 483–488.
- B. Xie, Z. Lv, B. Y. Lv, Y. X. Gu, Treatment of Mature Landfill Leachate by Biofilters and Fenton Oxidation, *Waste Manage.* **2010**, 30 (11), 2108–2112.
- Y. Y. Wu, S. Q. Zhou, D. Y. Chen, R. Zhao, H. S. Li, Y. M. Lin, Transformation of Metals Speciation in a Combined Landfill Leachate Treatment, *Sci. Total Environ.* **2011**, 409 (9), 1613–1620.
- F. Fikret, M. Y. Pamukoglu, Powdered Activated Carbon Added Biological Treatment of Pre-treated Landfill Leachate in a Fed-batch Reactor, *Biotechnol. Lett.* **2003**, 25 (9), 695–699.
- W. Zubot, M. D. Mackinnon, P. Chelme-Ayala, D. W. Smith, M. G. El-Din, Petroleum Coke Adsorption as a Water Management Option for Oil Sands Process-affected Water, *Sci. Total Environ.* **2012**, 427–428, 364–372.
- E. Soco, J. Kalemkiewicz, Adsorption of Nickel(II) and Copper(II) Ions from Aqueous Solution by Coal Fly Ash, *J. Environ. Chem. Eng.* **2013**, 1 (3), 581–588.
- S. Milicevic, T. Boljanac, S. Martinovic, M. Vlahovic, V. Milosevic, B. Babic, Removal of Copper from Aqueous Solutions by Low Cost Adsorbent-Kolubara Lignite, *Fuel Process. Technol.* **2012**, 95, 1–7.
- G. F. Wang, M. F. Bao, Z. Z. Han, Application Study on Heavy Metal Wastewater Treatment with Active Carbon, *Environ. Prot. Sci.* **2004**, 30 (2), 26–29.
- G. Issabayeva, M. K. Aroua, N. M. Sulaiman, Study on Palm Shell Activated Carbon Adsorption Capacity to Remove Copper Ions from Aqueous Solutions, *Desalination* **2010**, 262 (1–3), 94–98.
- R. Naseem, S. S. Tahir, Removal of Pb(II) from Aqueous/Acidic Solutions by Using Bentonite as an Adsorbent, *Water Res.* **2001**, 35 (16), 3982–3986.
- H. S. Shi, L. L. Kan, Leaching Behavior of Heavy Metals from Municipal Solid Wastes Incineration (MSWI) Fly Ash Used in Concrete, *J. Hazard. Mater.* **2009**, 164, 750–754.
- T. Mangialardi, Sintering of MSW Fly Ash for Reuse as a Concrete Aggregate, *J. Hazard. Mater.* **2001**, 87, 225–239.
- X. L. Hou, X. Q. Ma, Research on Disposing of Landfill Leachate Polluted by Heavy Metal Using Incineration Ashes of Burning Rubbish, *J. Subtrop. Resour. Environ.* **2007**, 2 (2), 30–36.
- American Society for Testing and Materials, ASTM C 618, Specification for coal fly ash and raw calcined natural pozzolan for use as a mineral admixture in Portland cement concrete, in *Annual Book of ASTM Standards*, American Society for Testing and Materials, West Conshohocken, USA **1994**.
- I. J. Alinnor, Adsorption of Heavy Metal Ions from Aqueous Solution by Fly Ash, *Fuel* **2007**, 86, 853–857.
- W. H. Choi, S. R. Lee, J. Y. Park, Cement Based Solidification/Stabilization of Arsenic-contaminated Mine Tailings, *Waste Manage.* **2009**, 29 (5), 1766–1771.
- H. A. Elliott, C. M. Denny, Soil Adsorption of Cadmium from Solutions Containing Organic Ligands, *J. Environ. Qual.* **1982**, 11, 658–662.
- H. C. Cho, D. Y. Oh, K. Kim, A Study on Removal Characteristics of Heavy Metals from Aqueous Solution by Fly Ash, *J. Hazard. Mater.* **2005**, 127, 187–195.
- N. Ünlü, M. Ersoz, Adsorption Characteristics of Heavy Metal Ions onto a Low Cost Biopolymeric Sorbent from Aqueous Solutions, *J. Hazard. Mater.* **2006**, 136, 272–280.
- X. L. Sun, Q. X. Zeng, C. G. Feng, Adsorption Kinetics of Chromium(VI) onto an Anion Exchange Fiber Containing Polyamine, *Acta Phys. Chim. Sin.* **2009**, 25 (10), 1951–1957.
- Y. S. Ho, G. McKay, Kinetic Models for the Sorption of Dye from Aqueous Solution by Wood, *Process Saf. Environ. Prot.* **1998**, 76 (2), 183–191.
- W. L. Sun, J. R. Ni, Applicability of Langmuir Equation to Copper Sorption by Loess, *Environ. Chem.* **2002**, 21 (1), 37–44.
- J. R. Bai, Q. Wang, H. Qjn, G. K. Zheng, Y. J. Yang, X. H. Guan, Kinetics and Thermodynamics of Adsorption of Heavy Metal Ions onto Fly Ash from Oil Shale, *J. Fuel Chem. Technol.* **2011**, 39 (5), 378–384.
- Y. C. Sharma, S. N. Umaa, S. Paras, F. Gode, Fly Ash for the Removal of Mn(II) from Aqueous Solutions and Wastewaters, *Chem. Eng. J.* **2007**, 132, 319–323.
- A. D. Papandreou, C. J. Stournaras, D. Panias, I. Paspaliaris, Adsorption of Pb(II), Zn(II) and Cr(III) on Coal Fly Ash Porous Pellets, *Miner. Eng.* **2011**, 24 (13), 1495–1501.
- B. Bayat, Comparative Study of Adsorption Properties of Turkish Fly Ashes, I. The Case of Nickel(II), Copper(II) and Zinc(II), *J. Hazard. Mater.* **2002**, 95 (3), 251–273.
- S. Chaiyasith, P. Chaiyasith, C. Septhum, Removal of Cadmium and Nickel from Aqueous Solution by Adsorption onto Treated Fly Ash from Thailand, Thammasat, *Int. J. Sci. Technol.* **2006**, 11, 13–20.
- Q. Xue, J. S. Li, Z. Y. Hu, Compound Stabilization/Solidification of MSWI Fly Ash with Trimercapto-s-triazine and Cement, *Water Sci. Technol.* **2012**, 66 (3), 689–694.